Multi-gap nodeless superconductivity in iron selenide $FeSe_x$: evident from quasiparticle heat transport

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The in-plane thermal conductivity κ of the iron selenide superconductor $FeSe_x$ ($T_c = 8.8$ K) were measured down to 120 mK and up to 14.5 T ($\simeq 3/4H_{c2}$). In zero field, the residual linear term κ_0/T at $T \to 0$ is only about 16 μW K^{-2} cm^{-1}, less than 4% of its normal state value. Such a small κ_0/T does not support the existence of nodes in the superconducting gap. More importantly, the field dependence of κ_0/T in $FeSe_x$ is very similar to that in $NbSe_2$, a typical multi-gap s-wave superconductor. We consider our data as strong evidence for multi-gap nodeless superconductivity in $FeSe_x$. This kind of superconducting gap structure may be generic for all Fe-based superconductors.

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Just as CuO-plane is the basic building block of high-T_c cuprate superconductors, the FeAs-layer is the basic structure of the newly discovered FeAs-based high-T_c superconductors.[1,2,3,4,5,6,7] The FeAs-layer consists of a Fe square planar sheet tetrahedrally coordinated by As. However, unlike the rigid CuO-plane in cuprates, partial substitution of Fe by Co or Ni, or As by P within the FeAs-layer can effectively induce superconductivity.[8,9,10,11,12] In this sense, the discovery of superconductivity in binary $FeSe_x$ ($T_c \simeq 8$ K) is of great interests, since it only contains the superconducting FeSe-layer which has identical structure as FeAs-layer, and the Se deficiency may cause the superconductivity.[13] More remarkably, the onset T_c can be enhanced to as high as 37 K for $FeSe_x$ under high pressure,[14,15,16] which further implies that superconductivity in $FeSe_x$ may have the same mechanism as in FeAs-based superconductors.

For this new family of high-T_c superconductors, the pairing symmetry of its superconducting gap is a key to understand the mechanism of superconductivity. Extensive experimental and theoretical work have been done to address this important issue for FeAs-based superconductors (for a theoretical review, see Ref. 17; for an experimental review, see Ref. 18). Although there is still no consensus, more and more evidences point to multi-gap nodeless superconductivity, possibly an unconventional $s^{\pm}$ paring mediated by antiferromagnetic fluctuations.[19] For the prototype $FeSe_x$ superconductor, however, there were very few experiments to study the superconducting gap structure. This is due to its relatively lower T_c and lack of sizable high-quality single crystals.[20,21] The measurements of in-plane magnetic penetration depth for polycrystal $FeSe_{0.85}$ are in favor of anisotropic s-wave superconducting gap or two gaps ($s + s$).[22] To clarify this important issue, more experimental work are needed for $FeSe_x$ superconductor.

Low-temperature thermal conductivity measurement is a powerful tool to study the superconducting gap structure.[23] The field dependence of the residual thermal conductivity κ_0/T for $BaNi_2As_2$ ($T_c = 0.7$ K) is consistent with a dirty fully gapped superconductivity.[24] For $Ba_{1-x}K_xFe_2As_2$ ($T_c \simeq 30$ K) and $BaFe_{1.9}Ni_{0.1}As_2$ ($T_c = 20.3$ K), a negligible κ_0/T was found in zero field, indicating a full superconducting gap.[25,26] However, $\kappa(T)$ was only measured in magnetic fields up to $H = 15$ T ($\simeq 1/4\ H_{c_2}$), thus can not show clearly whether the superconductivity has multi-gap character in FeAs-based superconductors.[25,26]

In this paper, we measure the thermal conductivity κ of a $FeSe_x$ single crystal with $T_c = 8.8$ K down to 120 mK and up to 14.5 T ($\simeq 3/4\ H_{c_2}$) to probe its superconducting gap structure. In zero field, κ_0/T is about 16 μW / K^2 cm, less than 4% of its normal-state value. Such a small κ_0/T should not come from the nodal quasiparticle contribution. It may simply come from the slight overestimation when doing extrapolation, due to the lack of lower temperature data. The field-dependence of κ_0/T is very similar to that in multi-gap s-wave superconductor $NbSe_2$. Based on our data, it is evident that $FeSe_x$ is a multi-gap nodeless superconductor.

$FeSe_x$ single crystals with nominal formula $FeSe_{0.82}$ were grown via a vapor self-transport method.[21] The ab-plane dimensions of as-grown crystals ranges from a few hundred μm to 1 mm. Energy Dispersive of X-ray (EDX) microanalysis (Hitachi S-4800) show that the actual Fe:Se ratio is very close to 1:1 in our $FeSe_x$ single crystals. The nominal formula $FeSe_{0.82}$ was used in the initial work by Hsu et al..[13] However, the actual superconducting phase was later determined to be $FeSe_{0.99\pm0.02}$ in Ref. 27 and $FeSe_{0.974\pm0.005}$ in Ref. 28. Therefore the EDX result of our $FeSe_x$ single crystals is consistent with these two later reports.

The ac magnetization was measured in a Quantum Design Physical Property Measurement System (PPMS). An as-grown single crystal with dimensions 1.0 × 0.40 mm^2 in the plane and 190 μm thickness along the c-axis was selected for transport study. Contacts were made directly on the sample surfaces with silver paint, which

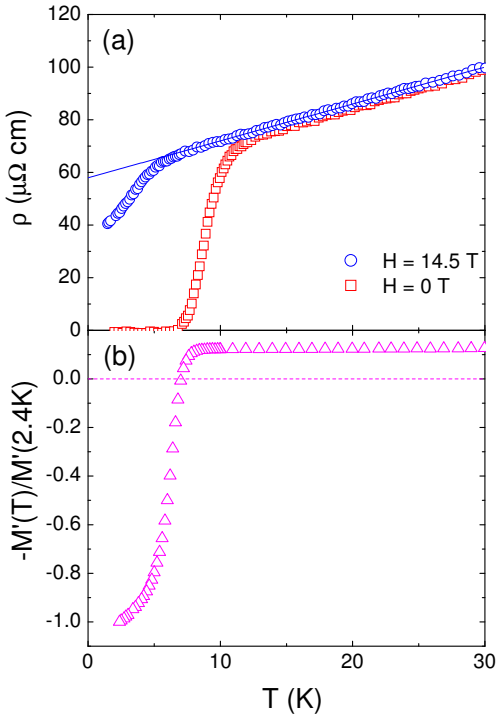

FIG. 1: (Color online) (a) In-plane resistivity $\rho(T)$ of $FeSe_x$ single crystal in $H = 0$ and 14.5 T magnetic fields along the c-axis. The solid line is a linear fit of $\rho(T)$ from 8 to 30 K, which gives the residual resistivity $\rho_0 = 57.9$ $\mu\Omega$ cm in $H = 14.5$ T. (b) Normalized ac magnetization.

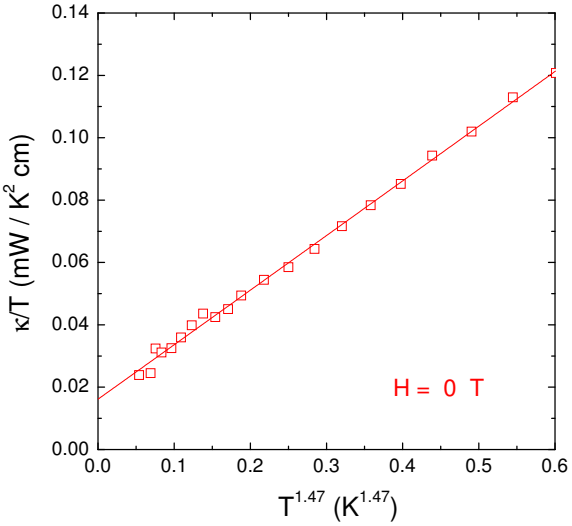

FIG. 2: (Color online) Temperature dependence of the in-plane thermal conductivity for $FeSe_x$ single crystal in zero field. The solid line represents a fit of the data to $\kappa/T = a + bT^{\alpha-1}$. This gives the residual linear term $\kappa_0/T = 16 \pm 2$ μW K^{-2} cm^{-1}.

were used for both resistivity and thermal conductivity measurements. The typical contact resistance is a few ohms at room temperature and 1.5 K, which is not as good as that on $Ba_{1-x}K_xFe_2As_2$ and $BaFe_{1.9}Ni_{0.1}As_2$ single crystals.[25,26] In-plane thermal conductivity was measured in a dilution refrigerator using a standard one-heater-two-thermometer steady-state technique. Due to the small size of the sample and the non-ideal contacts, good thermalization between sample and the two RuO_2 thermometers can only be achieved down to 120 mK. Magnetic fields were applied along the c-axis and perpendicular to the heat current. To ensure a homogeneous field distribution in the sample, all fields were applied at temperature above T_c.

Fig. 1a shows the in-plane resistivity of $FeSe_x$ single crystal in $H = 0$ and 14.5 T magnetic fields. The middle point of the resistive transition is at $T_c = 8.8$ K in zero field. The 10-90% transition width of our crystal is as broad as the powder sample,[13] which has been noticed in Ref. 21. Above T_c, $\rho(T)$ manifests a very good linear dependence up to 80 K, similar to the powder sample.[13] A linear fit of $\rho(T)$ gives the residual resistivity $\rho_0 = 57.9$ $\mu\Omega$ cm in $H = 14.5$ T, which is about 1/4 the value of powder sample.[13]

To estimate the upper critical field $H_{c2}(0)$ which completely suppresses the resistive transition, we define $T_c(onset)$ at the temperature where $\rho(T)$ deviates from the linear dependence, and get $T_c(onset) = 11.9$ and 6.3 K for $H = 0$ and 14.5 T, respectively. Using the relationship $H_{c2}/H_{c2}(0) = 1 - (T_c/T_c(0))^2$, we get $H_{c2}(0) = 20.1$ T. Note that $H_{c2}(0) = 16.3$ T was estimated for the powder sample, in which T_c was defined at the middle point of the transition.[13]

In Fig. 1b, the normalized ac magnetization for $FeSe_x$ single crystal is plotted. The positive ferromagnetic background has been attributed to the existence of Fe impurity in the $FeSe_x$ powder sample.[13] However, no iron, iron oxide, or iron silicide impurities were detected in our crystals,[21] therefore the ferromagnetic background likely results from the magnetic Fe cluster promoted by Se vacancies.[29]

Fig. 2 shows the temperature dependence of the in-plane thermal conductivity for $FeSe_x$ in zero field. To extrapolate the residual linear term κ_0/T, we fit the data to $\kappa/T = a + bT^{\alpha-1}$,[30,31] where aT and bT^{α} represent electronic and phonon contributions, respectively. In Fig. 2, the data from 120 mK to 0.7 K can be fitted (the solid line) and gives $\kappa_0/T = 16 \pm 2$ μW K^{-2} cm^{-1}, with $\alpha = 2.47$.

Such a value of κ_0/T is slightly larger than the experimental error bar ± 5 μW K^{-2} cm^{-1}.[31] However, it is still fairly small, less than 4% the normal state Wiedemann-Franz law expectation $\kappa_{N0}/T = L_0/\rho_0 = 0.423$ mW K^{-2} cm^{-1}, with L_0 the Lorenz number 2.45 $\times 10^{-8}$ W Ω K^{-2} and $\rho_0 = 57.9$ $\mu\Omega$ cm. For unconventional superconductors with nodes in the superconducting gap, a substantial κ_0/T in zero field contributed by the nodal quasiparticles has been found.[32,33] For example, for overdoped d-wave cuprate superconductor Tl2201 with $T_c = 15$ K, $\kappa_0/T = 1.41$ mW K^{-2} cm^{-1}, about 36% κ_{N0}/T.[32] For p-wave superconductor Sr_2RuO_4 with $T_c = 1.5$ K, κ_0/T

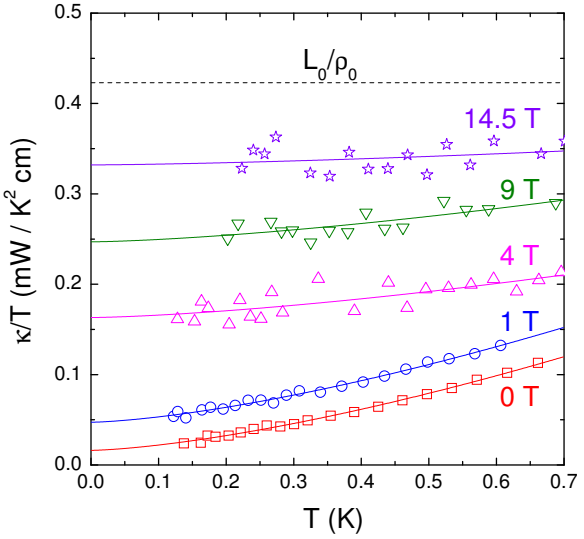

FIG. 3: (Color online) Low-temperature thermal conductivity of $FeSe_x$ in magnetic fields applied along the c-axis (H = 0, 1, 4, 9, and 14.5 T). The solid lines are $\kappa/T = a + bT^{\alpha-1}$ fits. For H = 4, 9, and 14.5 T, the electronic contribution becomes more and more dominant and the data get less smooth, therefore α is fixed to 2.47. The dashed line is the normal state Wiedemann-Franz law expectation at $T \to 0$, namely L_0/ρ_0, with L_0 the Lorenz number 2.45 $\times 10^{-8}$ W Ω K^{-2}.

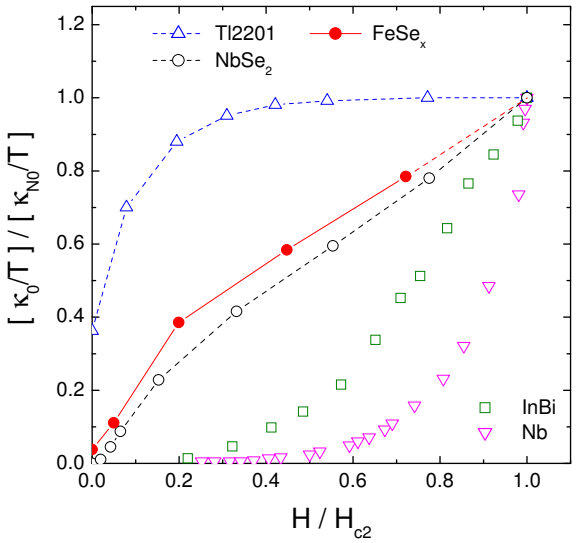

FIG. 4: (Color online) Normalized residual linear term κ_0/T of $FeSe_x$ plotted as a function of H/H_{c2}. Similar data of the clean s-wave superconductor Nb,[34] the dirty s-wave superconducting alloy InBi,[35] the multi-band s-wave superconductor $NbSe_2$,[36] and an overdoped sample of the d-wave superconductor Tl-2201[32] are also shown for comparison.

= 17 mW K^{-2} cm^{-1}, more than 9% κ_{N0}/T for the best sample.[33] We also note that κ_0/T in zero field are all negligible in closely related superconductors $BaNi_2As_2$, $Ba_{1-x}K_xFe_2As_2$, and $BaFe_{1.9}Ni_{0.1}As_2$.[24,25,26] Therefore, it is unlikely that $\kappa_0/T = 16 \pm 2$ μW K^{-2} cm^{-1} in $FeSe_x$ single crystal comes from the nodal quasiparticles. Since no impurity phases were detected, such a small κ_0/T may simply come from the slight overestimation when doing extrapolation, due to the lack of experimental data below 120 mK.

Below we turn to the field dependence of κ_0/T in $FeSe_x$. Fig. 3 shows the low-temperature thermal conductivity of $FeSe_x$ in magnetic fields applied along the c-axis (H = 0, 1, 4, 9, and 14.5 T). For H = 1 T, the data is also fitted to $\kappa/T = a + bT^{\alpha-1}$, and gives κ_0/T = 47 $\pm$ 2 μW K^{-2} cm^{-1}, with α = 2.47. For H = 4, 9, and 14.5 T, the electronic contribution becomes more and more dominant and the data get less smooth, therefore α is fixed to 2.47 in the phonon term bT^{α}. From Fig. 3, even higher magnetic field is needed to increase κ/T to its normal state value.

In Fig. 4, we put the normalized $\kappa_0/T(H)$ of $FeSe_x$ together with the clean s-wave superconductor Nb,[34] the dirty s-wave superconducting alloy InBi,[35] the multi-band s-wave superconductor $NbSe_2$[36] and an overdoped sample of the d-wave superconductor Tl-2201,[32] plotted as a function of H/H_{c2}. For a clean (like Nb) or dirty (like InBi) type-II s-wave superconductor with isotropic gap, κ_0/T should grow exponentially with field (above H_{c_1}). This usually gives negligible κ_0/T for field lower than $H_{c_2}/4$. For the d-wave superconductor Tl-2201, κ_0/T increases roughly proportional to $\sqrt{H}$ at low field due to the Volovik effect.[37] By contrast, for multi-gap superconductors $NbSe_2$ and MgB_2,[36,38] magnetic field will first suppress the superconductivity on the Fermi surface with smaller gap, and give distinct shape of $\kappa_0/T(H)$ curve, as seen in Fig. 4.

From Fig. 4, the $\kappa_0/T(H)$ of $FeSe_x$ manifests almost identical behavior as that of multi-gap s-wave superconductor $NbSe_2$. For $NbSe_2$, the shape of $\kappa_0/T(H)$ has been quantitatively explained by multiband superconductivity, whereby the gap on the Γ band is approximately one third of the gap on the other two Fermi surfaces.[36] Therefore, we consider our data as strong evidence for multi-gap nodeless superconductivity in $FeSe_x$. Note that in the two-gap $s + s$-wave model to describe the in-plane penetration depth data, the magnitude of the two gaps are 1.60 and 0.38 meV, respectively.[22] The ratio between these two gaps is about 4, close to that in $NbSe_2$, thus supports the multi-gap scenario from our thermal conductivity results.

So far, there is still no experiment to directly measure the superconducting gap in $Fe_{1+y}Te_{1-x}Se_x$ system. Density functional calculations show that the electronic band structure of FeS, FeSe, and FeTe are very similar to the FeAs-based superconductors.[39] In doped $BaFe_2As_2$, multi-gap nodeless superconductivity has been clearly demonstrated by angle-resolved photoemission spectroscopy (ARPES) experiments.[40,41,42] For hole-doped $Ba_{0.6}K_{0.4}Fe_2As_2$ (T_c = 37 K), the average gap values $\Delta(0)$ for the two hole pockets (α and β) are 12.5 and 5.5 meV, respectively, while for the electron (γ and

δ) pockets, the gap value is similar, about 12.5 meV.[40,41] For electron-doped $BaFe_{1.85}Co_{0.15}As_2$ (T_c = 25.5 K), the average gap values $\Delta(0)$ of hole (β) and electron (γ and δ) pockets are 6.6 and 5.0 meV, respectively[42]. The ratio between the large and small gaps is 2.3 for $Ba_{0.6}K_{0.4}Fe_2As_2$. This may explain the rapid increase of $\kappa_0/T(H)$ at low field in $Ba_{1-x}K_xFe_2As_2$,[25] although magnetic field was only applied up to 1/4 H_{c2} thus could not see clear multi-gap character as in our $FeSe_x$ single crystal.

In summary, we have measured the low-temperature thermal conductivity of iron selenide superconductor $FeSe_x$ to investigate its superconducting gap structure. A fairly small κ_0/T at zero field and the dramatic field dependence of κ_0/T give strong evidence for multi-gap nodeless superconductivity in $FeSe_x$. Such a gap structure may be generic for all Fe-based superconductors. More experiments are needed to distinguish conventional s-wave from the unconventional $s^{\pm}$-wave superconductivity in this new family of high-T_c superconductors.

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